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Sound Propagation in Bubbly Liquids. A Review

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CONTENTS

INTRODUCTION	1
EQUILIBRIUM SOUND SPEED	2
SOUND SPEED AT NON-ZERO FREQUENCIES	13
CONCLUSIONS	24
ACKNOWLEDGEMENTS	25
REFERENCES	25
APPENDIX A — Equilibrium Sound Speed in a Dilute Fluid-Particle Mixture	27
APPENDIX B — Radial Pulsations of Gas Bubbles in a Sound Field	39
APPENDIX C — Translational Motion of a Rigid Bubble in a Sound Wave	43

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SOUND PROPAGATION IN BUBBLY LIQUIDS. A REVIEW

INTRODUCTION

This work considers the propagation of sound waves in liquids containing bubbles. The problem has been given considerable attention in the past, but continues to be of interest, because it is of some importance in a variety of situations. For example, underwater waves used in surface-scattering experiments may encounter clouds of bubbles, whose effects on the waves must be determined if the scattered signal is to be correctly interpreted. Other examples include the study, by acoustic means, of bubbles created by the breaking of wind-driven surface waves.

Because the best known effect of the bubbles on the waves is to produce changes in the speed of propagation, there is a large number of works in the literature which deal with the speed of sound in a bubbly liquid. Unfortunately, the information available appears to lack the consistency required to provide a solid foundation for the study of different aspects of the problem. Further, the existing predictions for the speed of propagation appear to be based on *ad-hoc* assumptions which have not been critically examined. Consequently, their validity and limitations are not well established.

The focus of the work, then, is the speed of propagation in the simplest situation, namely, plane, monochromatic sound waves traveling in a large body of liquid containing small, non diffusing gas bubbles of uniform radius and devoid of surface tension effects. In particular, we consider the equilibrium and the finite-frequency speeds, as calculated by Wood (1941) and by Kennard (1943), respectively. In a separate report (included here as Appendix A), we show that Wood's equation is generally incorrect, but that in the case of bubbly liquids in water it gives correct numerical values because the specific heat of liquids is very close to unity, and because, for typical bubbly mixtures, the mass concentration of bubbles is a very small quantity. However, in some conditions, bubbles may act as rigid spheres, in which case the use of Wood's equation can produce significant errors, as

then, the speed of propagation differs little from the pure liquid value. Finally, In the case of finite frequencies, we conclude that Kennard's equation at frequencies near or slightly beyond radial resonance of the bubbles is questionable, at least for volume concentrations which are not exceedingly small.

EQUILIBRIUM SOUND SPEED

We first consider the propagation of sound waves in a bubbly liquid of infinite extent, when the bubble, or suspended phase, is in complete thermodynamic equilibrium with the host liquid. This situation occurs when the frequency of the sound waves is much lower than the lowest characteristic frequency of any mechanism playing a role in the propagation. An important example of such a mechanism is the radial resonance of the bubbles. Here the natural frequency in the absence of any dissipation, surface tension, and nonlinear effects is given by Minnaert's formula (Minnaert, 1933)

$$\omega_0 = \frac{c_p}{R_0} \sqrt{3\rho_p/\rho_f} \tag{1}$$

where R_o is the mean radius of the oscillating bubble, c_p is the speed of sound in the gas contained in the bubble, and ρ_p and ρ_f are the ambient densities of the gas and of the liquid, respectively. If this radial motion were the only effect induced by the waves, then we would require that the frequency of propagation ω be much smaller than ω_o . When other effects are present, we require that the frequency of propagation ω be much smaller than the lowest characteristic frequency associated with them. In such conditions, the two phases are in equilibrium with one another, so that we may use simple thermodynamic arguments to obtain the speed of sound in the mixture. Because thermodynamic equilibrium necessarily occurs at zero frequency, this equilibrium speed must therefore coincide with the zero-frequency limit, c(0), of a frequency-dependent speed of propagation, $c(\omega)$, which will be discussed in a later section. First, we consider some thermodynamic properties of the mixture and derive Wood's equation and the equilibrium speed of sound.

Equilibrium properties of a gas bubble-liquid mixture

We consider a unit volume of a mixture of gas bubbles and a liquid. For the mixture to be in equilibrium, it must be homogeneous. That is, every volume element in the mixture must contain similar distributions of bubbles. In such conditions, we may define define thermodynamic properties such as pressure, density, etc., in terms of the volume fractions of the bubble and liquid phases, and of the respective thermodynamic properties in the pure phases. We denote the volume fraction of the bubbles by C_v. The volume fraction for the liquid in the mixture is 1- C_v. Therefore, the density of the mixture is given by

$$\rho_{\rm m} = \rho_{\rm f}(1 - C_{\rm v}) + \rho_{\rm p}C_{\rm v} \tag{2}$$

Another quantity of interest is the mass loading, given by the ratio of gas mass to liquid mass in a unit volume. This is simply given by

$$\eta = \frac{\rho_p C_v}{\rho_f (1 - C_v)} \tag{3}$$

This quantity should not be confused with the mass concentration of bubbles in a unit volume of the mixture, Φ_p , which is given by

$$\Phi_{p} = \frac{\rho_{p}C_{v}}{\rho_{f}(1 - C_{v}) + \rho_{p}C_{v}} = \frac{C_{v}}{\delta(1 - C_{v}) + C_{v}}$$
(4)

where $\delta = \rho_{f}/\rho_{p}$. However, for very dilute suspensions, the two quantities are equal because $C_{v} << 1$, and $C_{v} << \delta$, giving

$$\Phi_{p} = \eta = C_{v}/\delta \tag{5}$$

For such suspensions, $\rho_m \sim \rho_f$.

Wood's equation

Perhaps the simplest formulation for the sound speed in a gross mixture may be found in the book by Wood (1941). It is based on the definitions of mixture density given earlier, and on a compressibility for the mixture, which Herzfeld (1930), Wood (1941), and others define as

$$K_{m} = K_{f} (1-C_{v}) + K_{p} C_{v}$$

$$(6)$$

Here K_f and K_p are the isentropic compressibilities of the liquid and and of the gas, respectively. Now, the isentropic compressibility of the liquid is given by

$$K_f = 1/\rho_f c_f^2 \tag{7}$$

and the isentropic compressibility of the gas is given by

$$K_p = 1/\rho_p c_p^2 \tag{8}$$

In terms of these quantities, Wood's compressibility is given by

$$K_m = (1 - C_v)/\rho_f c_f^2 + C_v/\rho_p c_p^2$$
 (9)

Thus, on the basis of these definitions, Wood introduces a low-frequency speed of propagation by means of

$$c_W^2 = \frac{1}{\rho_m K_m} \tag{10}$$

Using (2) and (9), we have

$$c_W^2 = \frac{1}{[(1-C_v)/\rho_f c_f^2 + C_v/\rho_p c_p^2][(1-C_v)\rho_f + C_v\rho_p]}$$
 (11)

This may be written as

$$\frac{c_f^2}{c_W^2} = (1-C_v)^2 + C_v^2 \frac{c_f^2}{c_p^2} + C_v(1-C_v) [N^2 + \rho_p/\rho_f]$$
 (12)

where

$$N^2 = \frac{\rho_f c_f^2}{\rho_p c_p^2}$$

is the ratio of liquid to gas compressibilities. For a water-air combination, N is about 124, so that we can safely discard the density ratio in the square bracket of the above equation. Finally, for the important case of dilute mixtures, this equation can be written as

$$\frac{c_f^2}{c_W^2} = \frac{1}{1 + C_V N^2} \tag{13}$$

Alternate derivation

Wood's equation was derived from first principles by Chambre (1954), who obtained it using three different, but equivalent, methods. For the case of gas bubbles in a liquid, the following variant of one of Chambre's derivation is equivalent to those found in the recent literature (see, for example, Batchelor, 1969; van Wijngaarden, 1972).

First, we define Wood's sound speed as

$$\frac{1}{c_{\rm w}^2} = \frac{d\rho_{\rm m}}{dp} \tag{14}$$

where p and ρ_m are the mixture's pressure and density, and where the derivative is to be evaluated at ambient conditions. Using Eq. (1), we obtain

$$\frac{1}{c_W^2} = \frac{1 - C_V}{c_f^2} + \frac{C_V}{c_p^2} + (\rho_p - \rho_f) \frac{dC_V}{dp}$$
 (15)

where

$$c_f^2 = \frac{dp}{d\rho_f} \tag{16}$$

$$c_p^2 = \frac{dp}{dp_p} \tag{17}$$

are the *isentropic* speeds of sound in the pure liquid and pure gas, respectively, also evaluated at ambient conditions.

The quantity dC_v/dp in Eq. (15) is not zero because the sound waves induce changes in the volume fraction. However, the mass concentration, or equivalently, the mass loading η , remains constant, because in equilibrium the two phases move together. We take advantage of this fact to evaluate that derivative, by using the isentropic equation of state for the gas, which states that, $p_p \, p^{-\gamma p}$ is a constant. (In doing so, we implicitly assume that all changes of state for the gas in the bubble are connected by isentropes.) Now, the mixture pressure p is, by the equilibrium assumption, uniform throughout the mixture. That is, it is the same in both liquid and gas phases. Therefore, $p_p = p$. This enables us to write Eq. (7) as

$$\frac{p^{-\gamma_p}C_v}{\rho_f(1-C_v)} = constant \tag{18}$$

Taking a derivative of this to form dC_{ν}/dp as needed, and evaluating it at ambient conditions, we find

$$\frac{dC_{v}}{dp} = C_{v}(1 - C_{v}) \left[\frac{1}{\rho_{f} c_{f}^{2}} - \frac{1}{\rho_{p} c_{p}^{2}} \right]$$
 (19)

Substitution into Eq. (14), yields Wood's equation.

Equilibrium sound speed

As the derivation above makes it clear, Wood's equation follows from the assumption that the mass concentration is constant. Such condition is, in fact, necessary for equilibrium. However, the equilibrium speed of sound in the mixture is defined as

$$c^{2}(0) = \left(\frac{dp_{m}}{d\rho_{m}}\right)_{S_{m}} \tag{20}$$

that is, at constant mixture entropy. In true thermodynamic equilibrium, the entropy of the mixture can remain constant only if the entropy increase of one of the components is matched with a decrease by the other. This is not the case with the derivation given earlier, which clearly requires only mechanical equilibrium. The question of thermal equilibrium, required for true thermodynamic equilibrium is not addressed in Chambre's derivations. Thus, in general, Wood's equation does not give the equilibrium sound speed in a gross mixture. Rather, as shown in a separate report included as Appendix A to this work, the correct value for the equilibrium, or zero frequency, sound speed is

$$c^{2}(0) = \frac{1 - \Phi_{p} \frac{(\gamma_{f}-1)}{1 - \Phi_{p}} (\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_{f}T_{0}})}{\rho_{m} [C_{v} K_{p} + (1 - C_{v}) K_{f}]}$$
(21)

where cpp and cpf are the specific heats at constant pressure of the

particles in the mixture and of the fluid, respectively, and γ_f is the ratio of specific heats for the fluid. This result may be written as

$$c^{2}(0) = \frac{1}{\rho_{m}K_{sm}}$$
 (22)

where we have introduced the isentropic compressibility of the mixture, K_{sm} , given by

$$K_{sm} = \frac{C_{v} K_{p} + (1 - C_{v}) K_{f}}{1 - (\gamma_{f} - 1) \frac{\Phi_{p}}{1 - \Phi_{p}} (\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_{f} T_{0}})}$$
(23)

Thus, in general, the isentropic compressibility of a gross mixture differs from that assumed by Wood (1941) and others. This may be significant is some situations. For example, in the case of droplets in a gas, the second term in the denominator of Eq. (22) can be of the order of one, even for dilute suspensions, so that Wood's equation will produce significant errors. However, in the case of bubbles in a liquid, that term is negligible because then, the mass concentration in suspensions of low volume concentration is very small, and because, for liquids, $(\gamma_f - 1) << 1$. For such a mixture, then, $K_m \approx K_{sm}$ and Wood's equation gives the correct numerical result for the speed of propagation at low frequencies. From now on, we will denote Wood's c_w by c(0).

Thus, the equilibrium sound speed in a bubbly liquid has a smaller value than in the host liquid alone, by an amount that depends on the volume concentration of the gas C_v . The dependence of $c^2(0)$ on C_v is shown in Fig. 1.

As C_v is varied from zero to one, the speed changes from the pure liquid value to the pure gas value, as expected, but for values of C_v in a wide range, it is even smaller than in the pure gas. At $C_v = 0.5$, for example, the speed of propagation is only about 20 m/sec, but, as pointed out by Batchelor (1969), it is difficult to imagine a homogeneous mixture of

bubbles in a liquid having such a high gas concentration.

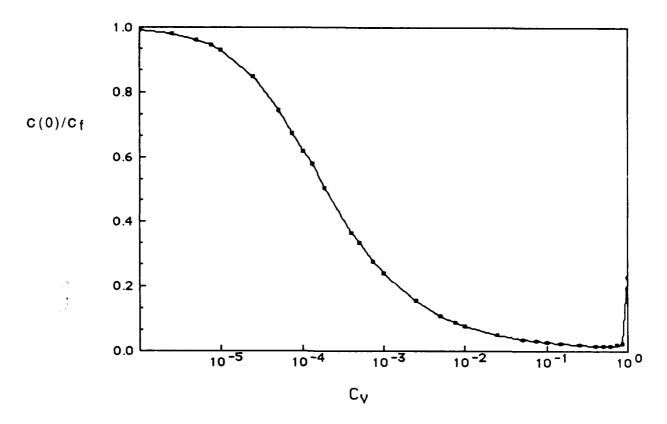


Fig. 1 Speed ratio $c(0)/c_f$ as a function of the volume fraction C_V . The value of $c(0)/c_f$ at $C_V = 1.0$ gives the ratio of speed in the pure gas to speed in the liquid.

In this work, we are interested only in mixtures having very small volume concentrations. However, even for these, the speed of propagation differs considerably from the value in the pure liquid. Thus, for example, Eq. (13) shows that a value of $C_{\rm V}$ as small as 1.85x10⁻⁴ decreases the propagation speed by 50%. Some of the consequences of such drastic changes will be examined later.

Rigid bubbles in a liquid

In some instances, bubbles in liquids behave nearly as rigid bodies due to the presence of surface impurities (see Batchelor, 1967). In these instances, Wood's equation can be used provided we put the bubble's compressibility equal to zero. Thus,

$$\frac{c_f^2}{c^2(0)} = (1 - C_v)^2 \approx 1 - 2C_v \tag{24}$$

Thus, in this case, the equilibrium speed of sound in the mixture is only slightly different from the speed in the pure liquid. This is important, for it predicts a much larger speed of propagation than that computed by assuming that the bubbles are clean, in which case we must use Eq. (13),

$$\frac{c_f^2}{c^2(0)} = 1 + C_v K_p/K_f$$
 (24a)

The ratio of fluid to liquid compressibilities appearing here is, for the case of air bubbles in water, larger than 16,000. Although the value of this ratio for actual bubbles found in practice depends on the amount of impurities on their surface, and this is generally not known, it is clear that a significantly lower value than 16,000 must apply in some cases, and this seems to have escaped the attention of previous investigators.

Some applications

Before concluding this section, we give some applications of the results presented in this section, assuming that we are dealing with clean bubbles.

1. Measurement of C_v

It should be noted that the theoretical derivation of Eq. (12) makes no assumption about the size of the bubbles. All that is assumed is that their distribution in the mixture is homogeneous, whether monodisperse (single size) or polydisperse. However, a direct measurement of C_v can sometimes be difficult to perform. On the other hand, speed of sound measurements are probably easier to make, and Eq. (13) offers an indirect manner of obtaining C_v . Thus, if a measurement of the speed of sound in the mixture is performed at very low frequencies, so that c_v is known, we may obtain c_v in a dilute mixture by means of

$$C_{v} = \left[\frac{c^{2}(0)}{c_{f}^{2}} - 1\right] N^{-2}$$
 (13a)

The procedure can also be used for non-dilute mixtures, but then the working equation is Eq. (12).

2. Reflection coefficient at liquid-mixture interface

It is known that in certain situations in the ocean, clouds of bubbles are found in the water as a result of breaking surface waves (Thorpe, 1982, 1987). If a sound wave were to meet such a cloud, a fraction of the incident energy may be scattered by the bubbly could, in a manner that depends on many factors, including cloud geometry, frequency, bubble size distribution, etc. An idea about the magnitude of the effect produced by the interaction, may be obtained by considering the reflection coefficient for plane waves of very low frequencies, normally incident in a bubbly cloud of semi-infinite extent. This is an application of the textbook example of the reflection at the interface between two infinite media, each having a different characteristic impedance. The well-known result for that situation is

$$\alpha_r = \left(\frac{\rho_f C_f - \rho_m C(0)}{\rho_f C_f + \rho_m C(0)}\right)^2 \tag{25}$$

where $c_{(0)}$ is, as before, the equilibrium speed in the mixture, and where the density of the mixture ρ_m is given by Eq. (2). For low volume concentrations, ρ_m is nearly equal to the speed of the liquid. Therefore, Eq. (23) reduces to

$$\alpha_{\rm r} = \left(\frac{C_{\rm f} - C(0)}{C_{\rm f} + C(0)}\right)^2 \tag{25a}$$

This result was apparently first given by Kennard in 1943. For finite volume concentrations, the dependence of α_r on C_v is shown in Fig. 2 below.

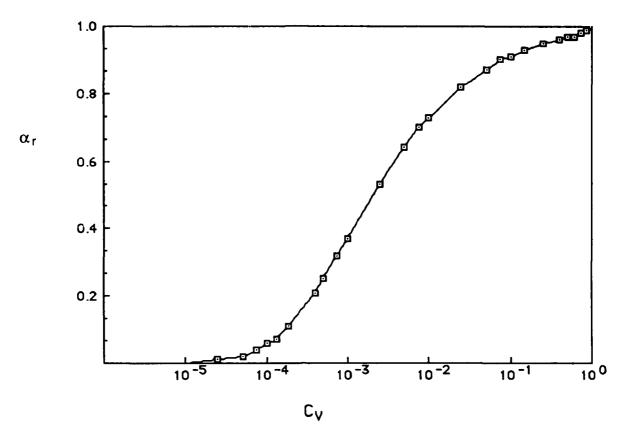


Fig. 2. Reflection coefficient at boundary between pure liquid and mixture.

These results show that a volume concentration equal to 1.85×10^{-4} , produces a reflection coefficient equal to 0.11. Thus, a non-negligible fraction of the incident energy is reflected back into the pure liquid, and this may be significant in some underwater scattering experiments. Incidentally, Eq. 25 may be used to obtain c(0) in terms of α_r . Thus,

$$\frac{c_f}{c(0)} = \frac{(1 + \sqrt{\alpha_r})}{(1 - \sqrt{\alpha_r})}$$
 (25b)

Hence, a measurement of the reflection coefficient made at low frequencies at an the interface between a bubbly and a pure liquid, will yield the speed ratio. This ratio can then be used to obtain C_{ν} .

SOUND SPEED AT NON-ZERO FREQUENCIES

As the discussion in the above section shows, sound propagation in bubbly liquids may take place at a significantly different speed than in the host liquid, even at very small volume concentrations of bubbles. The results presented above, however, are limited to equilibrium conditions in the mixture, and therefore can only be used at very low frequencies, although some authors indicate that the equilibrium speed of propagation predicted by Wood's equation can be used at moderate frequencies.

The literature contains several equivalent theories for the propagation, at finite frequencies, of sound waves in liquids with radially pulsating bubbles (see, for example, Carstensen and Foldy, 1947; Meyer and Skudrzyk, 1953; Drumheller and Bedford, 1979). These theories make it clear that the bubbles affect the propagation in a non-trivial manner which strongly depends on the bubbles response to the waves. In Appendices B and C, we will examine some of the aspects of this response. Here, we review the currently accepted theory for the finite-frequency speed of sound in a bubbly liquid. This theory is for liquids containing radially-pulsating gas bubbles. In this, as well as in other cases, propagation is dispersive, implying that the speed of propagation is frequency dependent, and that there is attenuation. In those conditions, the wavenumber k is complex, and may be expressed as

$$k = k_1 + i k_2$$
 (26)

where

$$k_1 = \omega/c(\omega)$$

$$k_2 = \alpha$$
(27)

where α is the *amplitude* attenuation coefficient, and ω is the circular frequency of the waves. Thus, a plane monochromatic wave propagating in the positive x-axis direction, produces in the bubbly fluid a pressure fluctuation proportional to $\exp(-\alpha x)\cos(k_1 x - \omega t)$

whereas in a pure liquid the equivalent result would be $cos(k_fx - \omega t)$, where $k_f = \omega/c_f$ is the wavenumber in the pure liquid. Of course, the problem is to determine k_1 and k_2 .

Perhaps the earliest investigation of the problem was that of Kennard (1943), who considered the effects of radially pulsating bubbles in an ideal liquid. His work is, however, not widely known, as it first appeared in a classified report. His results have been obtained by other investigators, on the basis of similar arguments, and may be found in standard textbooks on the subject (see, for example, Clay and Medwin, 1977). In what follows, we will first present them without derivation. A short derivation is presented after discussing some of the implications of the theory.

Now, in an ideal liquid, the dissipative effects of viscosity and heat conductivity are absent. Therefore, the only manner in which bubbles can remove energy from an incident acoustic wave, is by absorbing some energy from the wave, converting it into mechanical energy of radial pulsations, and radiating to infinity in the form of sound waves. This radiation is an energy loss as far as the incident wave is concerned, and results in attenuation and dispersion. Before writing Kennard's results, we introduce the relaxation time scale for radial pulsations defined by

$$\tau_{o} = \omega_{o}^{-1} \tag{28}$$

where ω_0 is Minnaert's equation for the natural frequency of an ideal bubble in the absence of surface tension effects. We call this the relaxation time scale for radial pulsations because it is the time scale associated with them. That is, significant changes in the acoustic variables occur during times that are comparable to τ_0 . With this definition, Kennard's results may be written as follows

$$\left(\frac{kc_{f}}{\omega}\right)^{2} = 1 + C_{v} N^{2} \frac{1}{1 - \omega^{2} \tau_{0}^{2} - i\left(\sqrt{3}/N\right) \omega^{3} \tau_{0}^{3}}$$
(29)

This may also be written as

$$\left(\frac{kc_f}{\omega}\right)^2 = X + i Y \tag{30}$$

where the real quantities X and Y are given by

$$X = 1 + C_{v} N^{2} \frac{1 - \omega^{2} \tau_{o}^{2}}{(1 - \omega^{2} \tau_{o}^{2})^{2} + ((\sqrt{3}/N) \omega^{3} \tau_{o}^{3})^{2}}$$
(31)

and

$$Y = C_{v} \sqrt{3} N \frac{\omega^{3} \tau_{o}^{3}}{(1 - \omega^{2} \tau_{o}^{2})^{2} + ((\sqrt{3}/N) \omega^{3} \tau_{o}^{3})^{2}}$$
(32)

Separating Eq. (30) into its real and imaginary parts, we have

$$\frac{c_f^2}{c^2(\omega)} - \overline{\alpha}^2 = X \tag{33}$$

$$2\overline{\alpha} c_f/c(\omega) = Y \tag{34}$$

where we have used the definitions of k_1 and k_2 given earlier, and where $\overline{\alpha}$ is a non-dimensional attenuation coefficient, defined by

$$\overline{\alpha} = \alpha c_f / \omega = \alpha \lambda / 2\pi \tag{35}$$

Equations 33 and 34 are readily solved in terms of the quantities X and Y defined earlier, giving

$$\frac{c(\omega)}{c_f} = \frac{1}{\left(\frac{1}{2}X + \frac{1}{2}\sqrt{X^2 + Y^2}\right)^{1/2}}$$
 (36)

and

$$\overline{\alpha} = \frac{Y/2}{\left(\frac{1}{2}X + \frac{1}{2}\sqrt{X^2 + Y^2}\right)^{1/2}}$$
(37)

These equations were first given by Kennard (1943) and are plotted as a function of $\omega \tau_0$ in Fig. 3, for air bubbles in water, and for a volume concentration $C_V = 1x10^{-4}$. We note that, according to these equations, and for a wide range of frequencies beyond resonance, the speed of

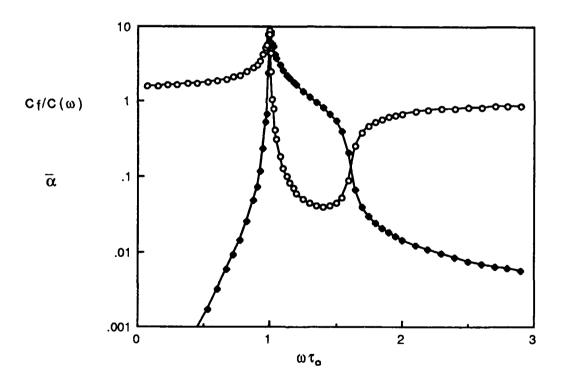


Fig. 3. Kennard's results for attenuation and dispersion. o: $c_f/c(\omega)$. $\phi: \overline{\alpha}$. $C_{V}=10^{-4}$.

propagation is significantly larger that the speed in the pure liquid. Also, the attenuation per wavelength exceeds unity in a wide frequency band. Of

course, for such large attenuations, one cannot properly talk about propagation, because the amplitude of the "propagating" waves decreases extremely rapidly. For example, the amplitude of a wave would decrease to less than 2/1000 of its initial value in a distance equal to $\lambda/2\pi$.

One reason for these remarkable features is that the amount of dissipation associated with radiation, represented by the second term in the denominator in Eq. (32) is very small. This has prompted some investigators to study additional dissipation mechanisms whose effects may decrease the maximum values of the attenuation. So that we can later introduce some other dissipative effects, we modify Kennard's equations using Minnaert's result, Eq. (1). This enables us to write

$$(\sqrt{3}/N) \omega^3 \tau_o^3 = k_f R_o \omega^2 \tau_o^2$$
 (38)

It is customary in the acoustics literature to write the quantity $k_f R_o$ in terms of an acoustic-radiation damping, and, in fact, several such damping coefficients have been introduced, which, as shown by the papers of Devin Jr (1959), Eller (1970), and Fairbank Jr (1975), have resulted in some confusion. To avoid further confusion, we will use the symbol 2β , as done by Prosperetti (1977), to represent damping. The precise definition of this quantity is made clear by simple reference to the bubble's equation for radial pulsations given in Appendix B. Thus, since in Kennard's case the damping is due to a monopole-type of radiation, we introduce a monopole radiation damping β_{rad}^m by means of

$$k_f R_o = 2\beta_{rad}^m \omega^{-1}$$
 (39)

As the defining equation shows, this damping coefficient is frequency dependent. With this notation, Kennard's basic equation is

$$\left(\frac{kc_{f}}{\omega}\right)^{2} = 1 + C_{v} N^{2} \frac{1}{1 - \omega^{2} \tau_{o}^{2} - 2i\beta_{rad}^{m} \omega \tau_{o}^{2}}$$
(40)

Because, as mentioned earlier, radiation damping produces very small dissipation, other damping coefficients have been introduced whose effects on the propagation coefficients are assumed to be correctly taken into account simply by replacing, in Eq. (40) $\beta_{\rm rad}^{\rm m}$ with

$$\beta = \beta_{rad} + \beta_{visc} + \beta_{ther} + \dots$$
 (41)

These separate coefficients are due to acoustic radiation, viscosity, heat transfer, etc., and may depend on the properties of the liquid, the gas, as well as on the frequency. We will now use this all-including damping for radial pulsations to derive Kennard's results.

Derivation of Kennard's equation

In its simplest form, the derivation of Eq. (40) is based on the equilibrium definition of the speed of sound, e.g.

$$\frac{1}{c^2} = \rho_m K_m \tag{42}$$

where, in view of our earlier comments, we have put $K_{sm} = K_m$. As in the equilibrium case, the compressibility of the mixture is now assumed to be functionally given by the sum of the liquid and gas compressibilities. Thus,

$$\left(\frac{k}{\omega}\right)^2 = \rho_m[(1 - C_v) K_{f+} C_v K_p] \tag{43}$$

where we have put $c(\omega) = \omega/k$. We now take the bubble concentration C_v to be very small so that the density of the mixture is basically equal to that of the liquid. The compressibility of the liquid at finite frequencies is taken to be equal to that of the pure liquid at zero frequency, i.e.,

$$K_f = 1/\rho_f c_f^2 \tag{7}$$

For the compressibility due to the gas bubbles in the liquid, we use the basic definition of that quantity

$$K_{p} = -\frac{1}{C_{vo}} \frac{C_{v} - C_{vo}}{P_{ac}}$$
 (44)

where P_{ac} is the acoustic pressure, assumed uniform around each bubble, and C_{vo} is the static value of C_v . Since $C_v = (4/3)n\pi R^3$, where n is the number of bubbles per unit volume of mixture, the changes in C_v , which are due to the radius of the bubbles changing from R_o to $(R_o + \epsilon)$, are approximately given by

$$C_{v} - C_{vo} = 3C_{vo}\varepsilon/R_{o} \tag{45}$$

where we have assumed that $|\epsilon/R_o| << 1$, and this in turn requires that $|C_v - C_{vo}|/C_{vo} \ll 1$.

The quantities P_{ac} and ϵ are given by Eqs. (B5) and (B7), respectively. Substituting these results in the above equation for K_p , we obtain

$$K_{p} = \frac{3}{\rho_{f}R_{o}^{2}} \frac{1}{\omega^{2} + 2i\beta \omega}$$

$$(46)$$

Returning now to our basic equation, we substitute these results to obtain

$$\left(\frac{kc_{f}}{\omega}\right)^{2} = \frac{\rho_{m}}{\rho_{f}} \left[1 + \frac{3C_{v}c_{f}^{2}}{\rho_{f}\omega_{o}^{2}R_{o}^{2}} + \frac{1}{1 - \omega^{2}\tau_{o}^{2} - 2i\beta\omega\tau_{o}^{2}}\right]$$
(47)

Using Minnaert's formula we have $\omega_o^2 R_o^2 = 3 c_p^2 \rho_p/\rho_f$. Finally, if we put $\rho_m = \rho_f$ we obtain

$$\left(\frac{kc_{f}}{\omega}\right)^{2} = 1 + C_{v} N^{2} \frac{1}{1 - \omega^{2} \tau_{o}^{2} - 2i\beta \omega \tau_{o}^{2}}$$
(48)

This is the same as Eq. (40), with β now taking a broader meaning.

Let us now examine this equation in some detail. First, we separate

its real and imaginary parts, to obtain

$$\frac{c_f^2}{c^2(\omega)} - \overline{\alpha}^2 = 1 + C_v N^2 \frac{1 - \omega^2 \tau_o^2}{(1 - \omega^2 \tau_o^2)^2 + (b\omega \tau_o)^2}$$
(49)

$$2\overline{\alpha} c_{V}/c(\omega) = C_{V} N^{2} \frac{b\omega \tau_{o}}{(1 - \omega^{2} \tau_{o}^{2})^{2} + (b\omega \tau_{o})^{2}}$$
 (50)

where $b = 2\beta \tau_i$ is a non-dimensional damping constant.

Except for the numerical coefficients appearing in these equations, they are the same as those which describe the well known phenomenon of "anomalous" dispersion of electromagnetic waves through an absorption band, and, in retrospect, it is not surprising to find that sound propagation through a liquid filled with radially pulsating bubbles of equal size, is described by the same equations. Now, the equations may be solved exactly for all values of the parameters appearing in them, and in fact, the solution is given by (36) and (37), with the quantities X and Y appearing in those equations now being given by

$$X = 1 + C_{v} N^{2} \frac{1 - \omega^{2} \tau_{o}^{2}}{(1 - \omega^{2} \tau_{o}^{2})^{2} + (b\omega \tau_{o})^{2}}$$
 (51)

$$Y = C_{v} N^{2} \frac{b\omega \tau_{o}}{(1 - \omega^{2} \tau_{o}^{2})^{2} + (b\omega \tau_{o})^{2}}$$
 (52)

This solution, with X and Y as above, will be referred to as the modified Kennard solution. Figure 4 shows the attenuation and speed of propagation as predicted by these equations, for the same C_v as that shown earlier, i.e., $C_v = 10^{-4}$, and for a non-dimensional damping constant equal to 0.1.

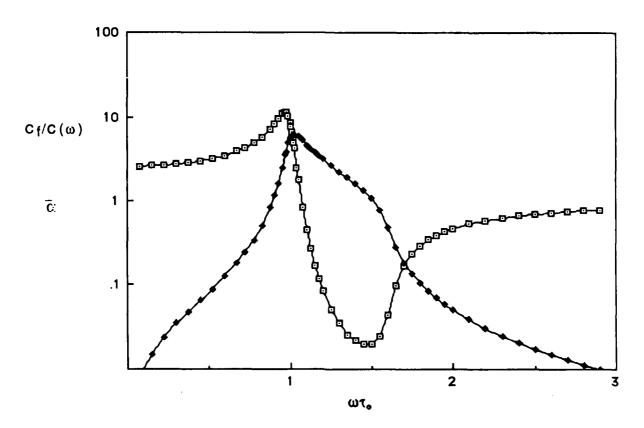


Fig. 4. Modified Kennard's attenuation and dispersion for $C_V = 10^{-4}$ and b=0.1 $\[\odot : c_f/c(\omega)$. $\[\bullet : \overline{\alpha} \]$

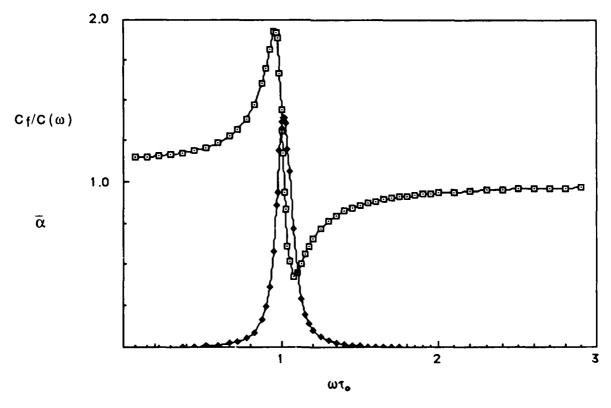


Fig. 5. Attenuation and dispersion for $C_V = 10^{-5}$ and b=0.1. \square : $c_f/c(\omega)$. \spadesuit : $\overline{\alpha}$

Figure 5 shows the same quantities for a volume concentration ten times smaller than that of the previous figure, but with the same damping constant. Figure 6 shows the same quantities for the same volume concentration as in Fig. 1, but with a damping constant ten times smaller.

It is clear from these figures that both damping and volume concentration have a most important effect on the propagation of sound waves in a bubbly mixture. We note, however, that regardless of how small the damping coefficient is, the typical curves (Fig. 5) associated with resonance occur only for the smallest values of C_v . Even a volume concentration of 10^{-4} produces significant distortion. It is also clear that the extreme values of speed and attenuation depend critically on both C_v and b, but the dependence is not simple except for very small volume concentrations. Nevertheless, several features stand out from these graphs that are worthy of notice. One of them is that the minimum value of $c_v/c(\omega)$ occurs for $\omega\tau_0 > 1$, and approaches zero as C_v is increased while holding b constant. A close examination of the exact solution shows that this limit is

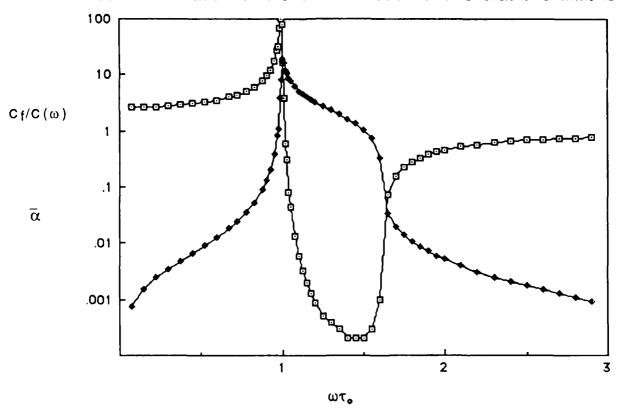


Fig. 6. Attenuation and dispersion for $C_V = 10^{-4}$ and b=0.01. \Box : $c_f/c(\omega)$. \spadesuit : $\overline{\alpha}$

never reached. However, a very small value of $c_f/c(\omega)$ implies a speed in the mixture which is considerably larger than that in the pure liquid. Such values of the speed ratio are, however, accompanied by non-dimensional attenuation coefficients which are of the order of one or larger. As remarked earlier, in connection with Kennard's original solution, such attenuation rates imply that sound waves do not propagate in the medium; they decay to negligible amplitudes in an extremely short distance, just as light waves are damped at an absorption band.

Let us now review some of the assumptions that we used in deriving this result, and which do not appear to have been examined before.

- 1. The damping coefficient β was taken to be a constant, and the effects of several distinct dissipative mechanisms were simply incorporated in the formulation by substitution of Eq. (41) into Eq. (40). The constancy of β is known to be incorrect, and the validity of the substitution is questionable.
- 2. The bubbly liquid may be described as a two phase medium. This assumption was used in ascribing properties to the mixture, and requires that there be large numbers of bubbles in every volume element. On the other hand, we have assumed that the pressure in the mixture is equal to that in the pure fluid alone, and this requires that the volume concentration be very small. In addition, we have computed a finite-frequency compressibility of the bubbles, using the results for a single bubble. This requires that the bubbles be sufficiently far apart from one another.
- 3. The changes in volume fraction must be small. Using Eq. (45), and Eq. (B8) this condition may be expressed as

$$\frac{|C_{v} - C_{vo}|}{|C_{vo}|} = \frac{P_{o}}{\gamma_{f} P} \frac{1}{(1 - \omega^{2} \tau_{o}^{2})^{2} + (2\beta \omega \tau_{o}^{2})^{2}}$$
 (53)

where P_0 is the pressure amplitude in the incident wave, and p is the mean pressure. Now, the amplitude of the sound wave is by assumption small

compared to the mean pressure. Thus, the above condition can be satisfied provided that the "amplification factor", given by the second fraction on the right hand side of the above equation, is, at most, of order one.

- 4. The compressibility of the mixture at finite frequencies is given by the same functional dependence on the individual-phase compressibilities as in the zero-frequency limit. This assumption has yet to be proven correct.
- 5. The bubbles are only pulsating radially. Appendix C considers the possibility of translational oscillation. Also, the possibility that surface impurities may cause the bubbles to behave as rigid spheres has not been considered.

CONCLUSIONS

This report has reviewed some of the characteristics of sound propagation in a bubbly liquid, giving special attention to the equilibrium sound speed, as given by the Wood equation, and to the relationships, first derived by Kennard, for the propagation constants at at finite frequencies, assuming that the changes are due to radially pulsating bubbles.

We conclude that although Wood's equation is generally incorrect, it gives the correct numerical value for the speed of propagation in a bubbly mixture in the limit of zero frequencies, at least for small volume concentrations of clean bubbles in liquids.

With regard to the modified Kennard's equations, we have shown that they produce the correct values for very large and very low frequencies, and that in between the values predicted are typical of wave propagation through an absorption band. It has also been pointed out that within that band, sound waves do not propagate in a bubbly medium, except for volume concentrations which are exceedingly small. At larger, but still very small concentrations, the waves are quickly absorbed. We have also pointed out some of the limitations of these results, and believe that some of the basic assumptions used in their derivation merit a further examination.

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APPENDIX A.

EQUILIBRIUM SOUND SPEED IN A DILUTE FLUID-PARTICLE MIXTURE

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INTRODUCTION

It is well known that the speed of sound in liquids containing bubbles is considerably different from the speed of sound in the liquids alone. The differences are produced by compressibility and density changes resulting from the presence of the bubbles. They can be very significant throughout the frequency range, and are particularly drastic at frequencies near the bubble's resonant frequency. However, even for frequencies that are very small compared to that resonant frequency, the speed of sound in the bubbly mixture can be considerably less than in the pure liquid. For example, a concentration, by volume, equal to 1.85 x 10⁻⁴, can decrease the speed at low frequencies by 50%. This can be very significant in several situations, particularly in low-frequency scattering experiments.

Because such differences are important, we have considered the speed of propagation for sound waves in a fluid containing small particles in suspension, in the limit of low frequencies, using equilibrium thermodynamic arguments. Particular examples include bubbles in a liquid, droplets in a gas, and other immiscible particles in a liquid or in a gas. We show that, in general, Wood's equation is incorrect, but that in the particular case of dilute suspensions of bubbles in water, the speed it predicts is numerically correct, owing to the low concentration of bubbles, by mass, and to the values of the thermodynamic properties of water. For other situations, significant differences can exist. In the case of bubbly liquids, we also point out that in the case of bubbles whose surfaces contain surface impurities, making them behave as rigid, the speed of propagation differs little from that in the pure liquid, and this may by

important. In the case of solid particles in a gas, our results agree with those that have been obtained in other studies.

2. WOOD'S EQUATION

In the limit of low frequencies, it is often assumed that the speed of propagation in a fluid-particle mixture is given by

$$c_W^2 = \frac{1}{\rho_m K_m} \tag{A1}$$

where

$$\rho_{\rm m} = \rho_{\rm p} C_{\rm v} + (1 - C_{\rm v}) \rho_{\rm f} \tag{A2}$$

is the density of the mixture, C_v is the volume concentration of the suspended particles and K_m is the compressibility of the mixture, which Wood (1931) defines in terms of C_v and of the compressibilities of the particles and of the host fluid. Thus,

$$K_{m} = K_{p}C_{v} + (1-C_{v})K_{f}$$
 (A3)

Now, the simplicity of Eq. (A1) is appealing, and in fact, equilibrium thermo-dynamics can be invoked to show that an equation of that type must exist in the limit of low frequencies. The problem is, however, that this compressibility is simply a model which has not been derived from first principles. It is true that Chambre (1954) has shown that Eq. (A3) is a logical consequence of the density law given by Eq. (2), but a cursory review of his work reveals that his K_m is not the isentropic compressibility of the mixture, as required by thermodynamics. Rather, his derivation of Eq. (A3) shows that Wood's K_m only requires that the mass concentration of the mixture be a constant. This condition insures mechanical equilibrium. It is therefore a necessary, but not sufficient condition, to insure thermodynamic equilibrium.

MIXTURE IN THERMODYNAMIC EQUILIBRIUM

The following derivation imposes thermodynamic equilibrium in the mixture, and assumes that the mixture is an ideal one. Effects such as adsorption or diffusion are not included. We will require some basic definitions. In addition to the quantities introduced above, we will need the mass concentration of particles, Φ_p , defined as the ratio of mass of particles to the mass of the mixture; the density of the particulate and fluid phases, σ_p and $\sigma_f = 1 - \sigma_p$, respectively. These are not to be confused with the densities of the particulate and fluid material, ρ_p and ρ_f , respectively. These quantities are interrelated in several ways, and the following relations among them will be used later.

$$\sigma_{p} = \rho_{p} C_{v} \tag{A4}$$

$$\sigma_f = (1 - C_v)\rho_f \tag{A5}$$

$$\Phi_{p} = \frac{\sigma_{p}}{\sigma_{p} + \sigma_{f}} = \sigma_{p}/\rho_{m} \tag{A6}$$

It follows from the above definitions that

$$\frac{1 - \Phi_p}{\Phi_p} = \frac{1 - C_v}{C_v} \delta \tag{A7}$$

where $\delta = \rho_f/\rho_p$. These enable us to write for the density of the mixture the equivalent, but more fundamental expression

$$\frac{1}{\rho_{\rm m}} = \frac{1 - \Phi_{\rm p}}{\rho_{\rm f}} + \frac{\Phi_{\rm p}}{\rho_{\rm p}} \tag{A8}$$

Also, from Eqs. (A2) and (A7) it follows that

$$\frac{\rho_{\rm m}}{\rho_{\rm f}} = \frac{1 - C_{\rm v}}{1 - \Phi_{\rm p}} \tag{A9}$$

Because the mixture is in equilibrium, we can define its entropy per unit mass as

$$s_m = (1 - C_v)s_f + s_p C_v$$
 (A10)

where s_f and s_p are the entropies, per unit mass, of the fluid and of the particles, respectively.

SPEED OF SOUND

We now proceed to obtain the equilibrium speed of sound in the mixture using the usual thermodynamic expression, i.e.,

$$c^{2}(0) = \left(\frac{dp_{m}}{d\rho_{m}}\right)_{S_{m}}$$
 (A11)

where p_m is the pressure in the mixture. This speed must correspond to the limit as $\omega \to 0$, of a frequency-dependent phase speed $c(\omega)$ in the mixture.

Now, we will assume that the total volume occupied by the particles is very small, and that the particles, while numerous enough to enable us to describe the medium as a homogeneous two phase medium, must nevertheless be separated by distances which are large when compared to their diameters. Under these conditions, the pressure in the mixture is simply equal to the pressure in the fluid alone, i.e., $p_m = p = p_f(\rho_f, s_f)$. Hence

$$dp_{m} = \left(\frac{dp_{f}}{d\rho_{f}}\right)_{S_{f}} d\rho_{f} + \left(\frac{dp_{f}}{ds_{f}}\right)_{O_{f}} ds_{f}$$
 (A12)

The first derivative on the right hand side is simply c_f^2 , the equilibrium sound speed in the pure fluid. The second derivative may be expressed as

$$\left(\frac{dp_f}{ds_f}\right)_{p_f} = \frac{p_f}{\beta_f} \left(\gamma_{f^-} 1\right) \tag{A13}$$

where β_f is the fluid's coefficient of thermal expansion and γ_f is its ratio of specific heats. Thus,

$$dp_{m} = c_{f}^{2} d\rho_{f} + \frac{\rho_{f}}{\beta_{f}} (\gamma_{f}-1) ds_{f}$$
 (A14)

Now, as Eq. (12) prescribes, the derivative there is to be taken at constant mixture entropy, which does not generally imply constant entropies for the mixture components. These are related by Eq. (A10), with $ds_m = 0$. Thus, because in equilibrium there is no relative motion between fluid and particles, Φ_p is a constant. Therefore, thermodynamic equilibrium requires that

$$ds_f = -\frac{\Phi_p}{1 - \Phi_p} ds_p \tag{A15}$$

This gives

$$dp_m = c_f^2 d\rho_f - \frac{\rho_f}{\beta_f} (\gamma_{f-1}) \frac{\Phi_p}{1 - \Phi_p} ds_p$$
 (A16)

Thus

$$c^{2}(0) = \frac{1}{\rho_{m}^{2}} \left[\frac{\Phi_{p}}{\rho_{p}^{2}} \frac{d\rho_{p}}{d\rho_{f}} + \frac{1 - \Phi_{p}}{\rho_{f}^{2}} \right]^{-1} \left[c_{f}^{2} - \frac{\rho_{f} (\gamma_{f} - 1) \Phi_{p}}{\beta_{f} (1 - \Phi_{p})} \frac{ds_{p}}{d\rho_{p}} \right]$$
(A17)

We now obtain approximate values for the total derivatives appearing in the right hand side of this expression. Consider first $d\rho_f/ds_p$. To compute its value, we note that the entropy change of the particles is

$$ds_p = c_{pp} \frac{dT_p}{T_p} - \frac{1}{\rho_p} \frac{dp}{T_p}$$
 (A18)

where dTp is the temperature change of the particles, and cpp is their

specific heat at constant pressure. Now, in equilibrium, dT_p must be equal to the fluid temperature change dT_f , and in an acoustic wave this is equal to

$$dT_f = \frac{\beta_f T_0 c_f^2}{\rho_f c_{pf}} d\rho_f \tag{A19}$$

Similarly, in equilibrium, the pressure is $dp = c_f^2 \, dp_f$. Further, the temperature changes are small so that in the denominator of we may put $T_p = T_0$. We thus obtain

$$\frac{ds_p}{d\rho_f} = \frac{\beta_f c_f^2}{\rho_f} \left(\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_f T_0} \right)$$
 (A20)

Substituting this result into EQ. (A17), yields

$$c^{2}(0) = \frac{1 - \Phi_{p} \frac{(\gamma_{f} - 1)}{1 - \Phi_{p}} (\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_{f} T_{0}})}{\rho_{m}^{2} \left[\frac{\Phi_{p}}{\rho_{p}^{2}} \frac{d\rho_{p}}{d\rho_{f}} + \frac{1 - \Phi_{p}}{\rho_{f}^{2}}\right]} c_{f}^{2}$$
(A21)

It remains to compute $d\rho_p/d\rho_f$. For the acoustic case, where the changes are due to an imposed pressure p', say, these are given by the isentropic relationships

$$d\rho_p = \rho_p K_p p', \qquad (A22a)$$

and

$$d\rho_f = \rho_f K_f p', \qquad (A22b)$$

where K_{p} and K_{f} are the isentropic compressibilities of the particles and of the fluid, respectively. Thus

$$\frac{d\rho_p}{d\rho_f} = \frac{\rho_p K_p}{\rho_f K_f} \tag{A23}$$

Substituting this into EQ. (A21), and using Eqs. (A7) and (A9), we obtain

$$c^{2}(0) = \frac{1 - \Phi_{p} \frac{(\gamma_{f}-1)}{1 - \Phi_{p}} (\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_{f}T_{0}})}{\rho_{m} [C_{v} K_{p} + (1 - C_{v}) K_{f}]}$$
(A24)

where we have used the definition of K_f to set $\rho_f c_f^2 K_f = 1$. This is the desired result. It can be put in the form of Eq. (A1) by defining the isentropic compressibility of the mixture as

$$K_{sm} = \frac{C_{v} K_{p} + (1 - C_{v}) K_{f}}{1 - (\gamma_{f} - 1) \frac{\Phi_{p}}{1 - \Phi_{p}} (\frac{C_{pp}}{C_{pf}} - \frac{\delta}{\beta_{f} T_{0}})}$$
(A25)

This differs from Wood's compressibility, Eq. (A3), by the factor

$$[1 - (\gamma_{f}-1) \frac{\Phi_{p}}{1 - \Phi_{p}} (\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_{f}T_{0}})]^{-1}$$

Thus, in general, Wood's equation is incorrect, being correct only when $\beta_f T_0 = \rho_f C_{pf}/\rho_p C_{pp}$, or when γ_f =1. However, for the specific case of gas bubbles in water, his equation yields the correct numerical value, because, then, both Φ_p and $(\gamma_f$ -1) are small. In other situations such as some of those considered below, differences may occur. To apply Eq. (A24) to special cases, it is useful to write it as

$$\frac{c_f^2}{c^2(0)} = \frac{(1 - C_v + C_v/\delta) (1 - C_v + C_v K_p/K_f) (1 - \Phi_p)}{1 - \Phi_p \left[1 + (\gamma_f - 1) \left(\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_f T_0}\right)\right]}$$
(A26)

BUBBLY LIQUIDS

Consider first the case when the particles are gas bubbles in liquids. Here, $\frac{\delta}{\beta_f T_0} >> \frac{c_{pp}}{c_{pf}}$, and $(\gamma_f - 1) \frac{\delta}{\beta_f T_0} >> 1$. Thus,

$$\frac{c_f^2}{c^2(0)} = \frac{(1 - C_v + C_v/\delta) (1 - C_v + C_v K_p/K_f)}{1 + \Phi_p (\gamma_f - 1) \frac{\delta}{\beta_f T_0}}$$
(A27)

However, in mixtures to which this work applies, the mass concentration is exceedingly small. Therefore

$$\frac{c_f^2}{c^2(0)} = 1 + C_v K_p/K_f$$
 (A28)

This is, of course, Wood's result for dilute bubbly suspensions.

RIGID PARTICLES

Consider now the case when the suspended particles may be regarded as rigid. This includes solid particles, small droplets, and may include small gas bubbles whose surfaces contain impurities making them appear as rigid. Of course, particles in a fluid may be considered rigid only if their compressibility, K_p , is much smaller than that of the surrounding fluid. For such cases, Eqs. (A2) and (A26) give

$$\frac{c_f^2}{c^2(0)} = \frac{(1 - C_v + C_v/\delta) (1 - C_v) (1 - \Phi_p)}{1 - \Phi_p \left[1 + (\gamma_{f^-}1) \left(\frac{c_{pp}}{c_{pf}} - \frac{\delta}{\beta_f T_0}\right)\right]}$$
(A29)

Two limiting situations are of particular interest.

1. $\delta <<$ 1. Dense particles in a gas

With $C_v = \Phi_p \delta <<1$, $\delta / \beta_f T_0 <<1$, we obtain

$$\frac{c_f^2}{c^2(0)} = \frac{1 - \Phi_p^2}{1 - \Phi_p[1 + \frac{C_{pp}}{C_{pf}}(\gamma_{f^-}1)]}$$
(A30)

For small mass concentrations, this yields

$$\frac{c_f^2}{c^2(0)} = 1 + \Phi_p \left[1 + \frac{c_{pp}}{c_{pf}} (\gamma_f - 1) \right]$$
 (A31)

This result agrees with those obtained earlier by Chu (1960) and by Rudinger (1965), who derived it for solid particles in a perfect gas. It also agrees with the limit of low frequencies of a more general result obtained by Temkin and Dobbins (1966).

2. $\delta >> 1$. Rigid bubbles in a liquid

Here, $\Phi_p = C_v/\delta << C_v <<$ 1. Hence,

$$\frac{c_f^2}{c^2(0)} = (1 - C_v)^2 \approx 1 - 2C_v$$
 (A32)

Thus, in this case, the equilibrium speed of sound in the mixture is only slightly different from the speed in the pure liquid. This is important, for it predicts a much larger speed of propagation than that computed by means of Eq. (A28). The ratio of fluid to liquid compressibilities appearing there is, for the case of air bubbles in water, approximately equal to 16,000. Although the value of this ratio for actual bubbles found in practice depends on the amount of impurities on their surface, and is generally not known, it is clear that a significantly lower value than the above figure must apply in some cases, and this seems to have escaped the attention of previous investigators.

3. Neutrally buoyant elastic particles

Finally, we consider the case when δ = 1. In this case, ρ_m = ρ_p = ρ_f , and C_v = Φ_p , so that Eq. (29) gives

$$\frac{c_f^2}{c^2(0)} = \frac{(1 - \Phi_p + \Phi_p K_p / K_f) (1 - \Phi_p)}{1 - \Phi_p \left[1 + (\gamma_{f^-} 1) \left(\frac{c_{pp}}{c_{pf}} - \frac{1}{\beta_f T_0}\right)\right]}$$
(A33)

This may be written as

$$\frac{c_f^2}{c^2(0)} = (1 - \Phi_p) \left[\frac{1 + \frac{\Phi_p}{1 - \Phi_p} \frac{K_p}{K_f}}{1 - \frac{\Phi_p}{1 - \Phi_p} (\gamma_{f^-} 1) \left(\frac{c_{p,p}}{c_{pf}} - \frac{1}{\beta_f T_0} \right)} \right]$$
(A33a)

For low mass concentrations, this yields

$$\frac{c_f^2}{c^2(0)} = 1 + \Phi_p \left[\frac{K_p}{K_f} + (\gamma_{f^{-1}}) \left(\frac{c_{pp}}{c_{pf}} - \frac{1}{\beta_f T_0} \right) \right]$$
 (A34)

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APPENDIX B

RADIAL PULSATIONS OF GAS BUBBLE IN A SOUND FIELD.

We consider here the radial pulsations of a small gas bubble due to an imposed monochromatic sound field of circular frequency ω . Although the bubble's spherical shape is due to surface tension effects, we will not include them, even though they may be important for some range of bubble sizes. In the linear approximation, the bubble behaves as a harmonic oscillator, so that the departures ϵ from the equilibrium radius R_0 are small, and are described by

$$M_o \ddot{\varepsilon} + 2\beta M_o \dot{\varepsilon} = -4\pi R_o^2 (p_b - p_e)$$
 (B1)

where p_b is the pressure in the bubble, p_e is the pressure outside it, M_o is the added mass of the bubble, and where β is the damping coefficient, assumed to represent all dissipative effects acting as a result of the radial pulsations. Now, the external pressure is equal to the sum of the equilibrium pressure p_a , and the acoustic pressure p_a . If the wavelength of the sound wave is very large compared to the equilibrium bubble radius, we may take the pressure around the sphere to be uniform, so that every portion of its surface is acted by a radially-directed force of equal magnitude. Similarly, we take the pressure inside the bubble to be uniform an equal to the equilibrium pressure p_a , plus the excess pressure due to small changes in bubble size. We take those changes to occur isentropically, which implies that the imposed frequencies cannot be too high. In these conditions,

$$p_b - p_o = -3\gamma_p p \varepsilon / R_o$$
 (B2)

where γ_{p} is the ratio of specific heats of the gas in the bubble. This gives

$$\ddot{\varepsilon} + 2\beta \dot{\varepsilon} + \omega_0^2 \varepsilon = -4\pi R_0^2 M_0^{-1} P_{ac}$$
 (B3)

where ω_b is Minnaert's resonant frequency. Because we have assumed that the pressure is uniform around the bubble, we must limit these results to frequencies such that $k_f R_o << 1$. In this limit, the added mass is equal to $4\pi R_o^3 \rho_f$. Now, for the acoustic pressure, we assume that the incident acoustic field is a plane wave, described by a potential ϕ given by

$$\phi = Ae^{i(k_f x - \omega t)}$$
 (B4)

The acoustic pressure around the sphere is obtained from this potential by the usual acoustic relation $P_{ac} = -p_f(\partial \phi/\partial t)$. This gives

$$P_{ac} = i\omega \rho_f A e^{-i(\omega t - kx)}$$
 (B5)

At x=0, where the bubble is located, this gives

$$\ddot{\varepsilon} + 2\beta \dot{\varepsilon} + \omega_0^2 \varepsilon = -i\omega A e^{-i\omega t} / R_0$$
 (B6)

Assuming a solution of the form

$$\varepsilon = \operatorname{Re}\left(\varepsilon_{o} e^{-i\omega t}\right) \tag{B7}$$

we obtain

$$\tilde{\varepsilon}_{0} = -\frac{i\omega A}{R_{0}} \frac{1}{\omega_{0}^{2} - \omega^{2} - 2i\beta \omega}$$
 (B8)

The radial velocity of points on the bubble surface is obtained from $U_b^r = (d\epsilon/dt)$. Thus, if we write

$$\tilde{U}_{b}^{r} = \tilde{U}_{bo}^{r} e^{-i\omega t}$$
 (B9)

then

$$\widetilde{U}_{bo}^{r} = -\frac{\omega^{2}A}{R_{o}} \frac{1}{\omega_{o}^{2} - \omega^{2} - 2i\beta \omega}$$
(B9)

This gives

$$\tilde{U}_{b}^{r} = -\frac{\omega^{2}A}{R_{o}} \frac{e^{-(i\omega t - \eta)}}{[(\omega_{o}^{2} - \omega^{2})^{2} + (2\beta\omega)^{2}]^{1/2}}$$
(B10)

where

$$\tan \eta = \frac{2\beta\omega}{\omega_0^2 - \omega^2}$$
 (B11)

APPENDIX C

TRANSLATIONAL MOTION OF A RIGID BUBBLE IN A SOUND WAVE

Because the size of the bubbles is usually considerably smaller than the wavelength of the sound waves, it is usually assumed that the pressure around the bubble is uniform. That is, if a bubble is placed in a plane sound wave that has a pressure distribution given by $P_o exp(ikx - \omega t)$, the pressure around the bubble is given by the sum of the ambient pressure, p, plus a time dependent pressure, given by $P_o exp(-i\omega t)$. This is deemed to be a valid approximation because on the bubble's surface, the spatial dependence varies by an amount which is of the order of kR_o , and this is negligible in the conditions given. However, the presence of a bubble necessarily modifies the acoustic field, making it not completely uniform around the bubble. Such a nonuniformity would induce the bubble to move in the direction of the wave, that is, to execute translational oscillations. As it may be anticipated, the most significant modifications occur when the effects of viscosity are taken into account, for then the bubble must adhere to the surrounding fluid.

To compute the magnitude of the variations of pressure around the bubble, we simply use an order of magnitude argument based on well-known results. A more detailed description of the translational response of a rigid bubble to a sound field may be found elsewhere (Temkin & Leung, 1977). Now, in the immediate vicinity of the bubble, the fluid can be regarded as incompressible. Therefore, provided the frequency and the Reynolds number are very small, the pressure in the immediate vicinity of the surface of the bubble, differs from its "free stream" value by an amount that is proportional to $\mu_f(U_0/R_0)$ cos θ , where μ_f is the viscosity coefficient of the liquid around the bubble, and θ is the polar angle between the position vector and the instantaneous fluid velocity, measured from the forward stagnation point, and Uo is the velocity amplitude in the wave. The magnitude of this pressure is small compared to the acoustic fluid pressure, pfcfUo, which exists without the bubble. However, it and the accompanying shear stresses produces a lateral force that induces a lateral motion. The effects of such a motion on the speed of propagation have not been investigated, although Batchelor (1969) considered its as an additional damping mechanism. However, it is clear that this translational motion adds a relaxation mechanism which is capable of inducing further changes in the speed of propagation.

Derivations of the equations for the translational oscillations of a rigid sphere in a sound wave travelling in a viscous fluid were presented elsewhere (Temkin and Leung, 1976). Here we simply reproduce the basic results for the case when the sphere density is much smaller than that for the fluid, and for the case when the force on the sphere can be computed from incompressible fluid theory. These results are approximately valid for small bubbles, because they appear to behave as rigid spheres owing to impurities in the host liquid which contaminates the interface.

Now, under the influence of the sound wave, the bubble will execute translational oscillations in the direction of the wave. Its translational velocity may be expressed as

$$U_{b}^{t} = U_{bo}^{t} e^{-i\omega t}$$
 (C1)

Because the bubble is much less dense than the surrounding fluid, it is convenient to introduce a dynamic translational relaxation time for the bubble, τ_{db} , defined by

$$\tau_{\rm db} = R_{\rm o}^2 / 9 \, v_{\rm f} \tag{C2}$$

The meaning of this relaxation time may be easily grasped by reference to the equation of motion of a rigid bubble, This may be expressed as

$$m_b(1 + \frac{1}{2} \frac{\rho_w}{\rho_a}) \frac{dU_b^t}{dt} = 6\pi \mu_f R_o(U_b^t - u) + ...$$
 (C3)

where the dots represent additional forces. Here \boldsymbol{U}_{b}^{t} is the translational

velocity of the bubble, m_b is its mass, and u is the fluid velocity. As already stated, this equation assumes that the bubble is rigid. Now, because the density of the liquid is much larger than that of the gas of the bubble, we have, approximately, $m_b(1+\frac{1}{2}\frac{\rho_w}{\rho_g})=(2/3)\pi R^3\rho_f$. The bubble's equation of motion can therefore be written as

$$\frac{dU_{bb}^{t}}{dt} = (U_{b}^{t} - u)/\tau_{db} + \dots$$
 (C4)

This is a typical relaxation equation, so that τ_{db} , as defined above, is seen to play the role of the relaxation time for the translational motion. For future reference, we note that this time scale is related to the radial relaxation time τ_0 by means of

$$\tau_{db} = \sqrt{3\rho_p/\rho_f} \frac{R_o c_p}{9v_w} \tau_o$$
 (C5)

For a 0.01 cm air bubble in water, this gives $\tau_{db}=10^3~\tau_{o.}$ Thus, the frequencies where translational relaxation effects are most significant, i.e., when $\omega\tau_{db}\sim 1$, correspond, for this radius, to a value of $\omega\tau_{o}$ equal to about 0.001. Again, for the same bubble size, the most significant translational effects occur at a frequency in the vicinity of 140 Hz. The magnitude of this effect is not known exactly, but as Batchelor (1969) points out, the changes it produces on the speed of propagation are likely to be small.

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